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by

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ABSTRACT

The Division of Nuclear Fuel Cycle of the International Atomic Energy Agency requested the assistance of CANMET in the preparation of a manual on the analytical chemistry in the uranium industry. The staff of the Chemical Laboratory has responded to this request and this report is intended to be part of the CANMET contribution to the analytical chemistry manual. This report deals with the determination of radon in the field.

MÉTHODES POUR MESURER LES CONCENTRATIONS DE RADON SUR LE TERRAIN

par

R. McCorkell

RÉSUMÉ

La Division du cycle du combustible nucléaire de l'Agence internationale de l'énergie atomique a demandé au CANMET de participer à la préparation d'un manuel de chimie analytique à l'intention de l'industrie de l'uranium. Le personnel du Laboratoire de chimie a répondu à cette demande, et le présent rapport fait partie de la contribution du CANMET au projet. Ce rapport traite de la détermination du radon sur le terrain.

INTRODUCTION

The measurement of radon concentrations or activities in the field has been covered exhaustively by Dyck and Jonasson (1). The present article is an abridgement of the relevant sections of the above reference; style and contents were retained where possible.

All current Rn detection methods depend on the radioactive properties of Rn and its decay products. With the invention of the photo multiplier in 1940, activated ZnS screens became the dominant Rn detectors. The introduction of integrated circuits and microprocessors had led to the development of compact sophisticated instruments. Several articles describing one or more methods are in print; therefore this discussion will deal only briefly with the principle of each method and refer the reader to original publications for more details. For convenience, the methods are subdivided into instantaneous, semi-integrating and fully-integrating categories. By 'instantaneous' is meant that an answer is obtained within minutes. Semi-integrating methods give an answer in several hours or days and offer the advantage that they integrate or smooth out diurnal fluctuations. Fully integrating techniques can average Rn signals for several weeks or months.

Principles of Methods

When a Ra atom decays, it produces an alpha particle and a radioactive daughter, Rn, which in turn decays. Thus between ^{222}Rn and ^{210}Pb there occur 3 alpha decays and 2 beta decays within a reasonable short time span compared to the half lifes of ^{226}Ra and ^{210}Pb. The alpha particle ejected from a disintegrating atom dissipates its energy by colliding with matter and ionizing atoms and molecules in its path. When the ionization takes place in activated ZnS, flashes of light in the visible region are produced. The activation is achieved by introducing an impurity, usually Ag, into the lattice of ZnS giving rise to electron transition bands which emit energy or photons when excited, which can be detected and recorded.

A solid state or surface barrier detector uses the energy expended by the α-particles at the p-n junction of a silicon diode.

The thermoluminescent dosimeter uses a phosphor, such as $CaSO_4$ doped with Dy, with the ability to trap electrons between the conducting and the valence bands. These trapped electrons will return quickly to the valence band when the phosphor is heated, producing a light pulse than can be recorded with a photomultiplier tube.

When an alpha particle passes through matter such as a cellulose nitrate film it leaves a trail of broken molecular bonds. Such tracks are enlarged to sizes visible through a microscope by etching the surface of cellulose nitrate with an aqueous solution of NaOH. These are called etched track detectors.

Each of the above principles can be used in one or more of an instantaneous, semi-integrating or fully-integrating mode. Several reviews of methods, instruments, and applications have been published (2,3,4,5,6,7).

The alpha scintillometer. There are probably more versions of the alpha scintillometer in use than any other Rn detection device. When Rn gas is brought into the ZnS(Ag) cell it is commonly referred to as the Rn sniffer. The reason for its popularity is its relative simplicity, ruggedness, versatility, sensitivity and low cost. Rn concentrations of a Bq/L in gases or waters can be measured in 10 minutes without appreciable interference from humidity, dust or temperature changes. There are many accounts of alpha particle detection instruments for the measurement of Rn (8,9,10,11,12,13,14,15).

Semi-integrating mode

Two methods lend themselves to this mode of Rn determination: the "Collector" and the "ROAC" (Rn on activated charcoal) methods.

(a) The collector method is one of the most recent innovations in U exploration instrumentation (16,17) even though the principal has been employed in Rn studies for decades. It is based on the collection and detection of the decay products of Rn. When a clean surface, such as a metallic or plastic disc is exposed to an atmosphere containing Rn, the decay products of Rn will collect on the disc. The activity of these daughters builds up with a composite half life of about 35 min and reaches a maximum in about 3¼ h.

There is a wide range of detectors to choose from. One of the simplest and most sensitive is the alpha scintillometer described by Card and Bell (16). It has also been applied to soil samples in the laboratory (16,18). The collector is suspended from an inverted cup, which is in a hole, covered with a plastic blanket and soil and left for 4 hours or more. Then the collector is transferred to the ZnS(Ag) coated chamber of the alpha scintillometer and its alpha activity determined. A recent novel commercial innovation of the collector method is the alphaCard of AlphaNuclear Ltd., Toronto, Canada. It uses a plastic film in a slide for the collector and two solid state detectors facing each other inside a portable computerized alpha counter.

A novel and efficient variant of the collector method was patented by Powell (19,20). The decay products of Rn are collected on a piece of polyester film suspended at the top of an aluminum tube inserted in the ground.

(b) The radon on activated charcoal (ROAC) method. The fact that Rn adsorbs on activated charcoal has been used for a long time (21,22,23). By burying opened charcoal cartridges contained in cups the system can be used in the semi-integrating mode. Because it takes time for Rn to diffuse from soil into the cartridge, 8 to 10 days are needed to saturate the charcoal. Hence signal integration is in the order of days and diurnal variations are smoothed out.

Fully-integrated mode

Photographic and etched cellulose nitrate films and solid state detectors with built-in pulse registers are fully-integrating mode methods. They accumulate and integrate alpha pulses as long as they are exposed to alpha activity. Three proprietary integrating techniques, Track-Etch, the alphameter and the Thermoluminescent Dosimeter came onto the market in the 1970's.

(a) The Track-Etch Method. This is probably the most widely publicized method for prospecting (2,24,25). The film, placed in a plastic cup, is placed in site for 3 or 4 weeks recovered and etched in NaOH solution to make visible the tracks left by the impinging alpha particles. The tracks

per unit area are counted under a microscope. Tracks from thoron can be prevented by placing a thin plastic film over the cups (26,27).

(b) The alphameter method became practical when inexpensive silicon diodes and integrated circuits were developed. Alpha particles striking the active surface of the diode are counted and stored until a reader is connected and the accumulated count recorded (28,29).

(c) Thermoluminescent dosimeters. These are a recent addition to Rn methods of prospecting (30). As with the other integrating devices, the thermoluminescent device (TLD), usually a protected thin wafer of Dy-doped $CaSO_4$, is placed inside a cup and buried in the soil for a set time. Alpha particles deposit all their energy in it, causing a proportional number of electrons to be trapped in intermediate energy states or bonds.

When the TLD is heated, the electrons return to the groundstate or valence band and the energy release at a luminescence centre of the phosphor causes visible light to be emitted the intensity of which is proportional to the initial alpha particle energy.

Determination of Rn in natural waters

Methods of determining Rn in natural waters usually involve the removal of Rn from an aliquot of sample by passing a fine stream of air bubbles through it, filling an evacuated ZnS(Ag) cell with this air-Rn mixture and counting the alpha particle activity in the cell with an alpha scintillometer. The parts can be interconnected with metal and plastic tubing. Use of rubber or plastic tubing should be minimal; Rn is absorbed by rubber. Consequently, it may take hours to clean a rubber tube which has been exposed to a very active sample to minimize the risk of cross-contamination of more normal samples. The fritted disc at the bottom of the sample holder disperses air into fine bubbles resulting in more efficient scrubbing of the water sample. The cell is evacuated with a hand or electrically operated pump and air is allowed to flow back in through the fritted disc and water sample.

The errors (associated with the method) can be divided into four types: instrumental, experimental, sampling, and statistical. Instrumental errors include detection efficiency changes of cells and photomultipler, changes in the gain of amplifiers, high voltage drifts, and faulty operation of other components such as pressure gauges, timers, and valves. The main experimental errors are pressure, volume, and time measurements, and the degassing procedure. Sampling errors will result mainly from the escape of radon due to its high dispersivity. To minimize these, glass bottles with well-sealing caps should be used and filled to the top except for one air bubble. This bubble is necessary to prevent the tightly sealed bottle from cracking during temperature changes. The largest uncertainty in the radon values results from the inhomogeneous distribution of radon in natural water bodies. Experience has shown that the act of sampling can disturb the water and change the radon concentration at the site by as much as a factor of 5. In view of such large variations, the other errors including the errors associated with the randomness of radioactive decay, become negligible. With constant radon sources the statistical counting error is dominant in all but very active samples (several Bq or more).

It should be noted that the same method can be used for the determination of Ra in natural waters, simply by degassing a sample completely and letting the Rn grow back in for a known length of time in a sealed bottle. The same technique may also be used for the determination of Ra in rocks and soils. The solid samples are dissolved and the solutions treated in the same manner as the water samples. Equivalent U can be calculated from the Ra concentration using the expression

$$eU(ppm) = 80.05 \times Ra(Bq/g)$$

The constant embodies the half lifes of $^{238}U(4.468 \times 10^9)$ and $^{226}Ra(1600\ y)$ and allows for 0.7% ^{235}U in natural U.

Determination of Rn in soil gas/radon emanations

Semiquantitative determinations of Rn in soil air are carried out relatively quickly with the alpha scintillometer. In the dynamic mode soil gas is pumped from a hole through the detector to the atmosphere and a Rn reading taken when the signal stabilizes. In the static mode soil gas is

circulated through the hole and detector in a closed loop. A great variety of soil gas probes have been developed for different soil types and applications. However, all probes have one common deficiency, they are difficult to hammer into the ground to a depth of 50 cm or deeper. The Rn content of soil air, for a variety of reasons, varies most in the top 50 cm. To minimize environmental and depth variations, it is advisable to go to a depth of at least 100 cm. At these or greater depths the Rn flux becomes progressively more stable and quantitative determinations can be made. Errors caused by the presence of thoron can be avoided evacuating a vacuum tight container (or cell if sufficient cells are available) with a high quality hand pump, letting the pure soil air from a probe in the ground flow into the cell and taking the filled cells to a field laboratory for counting and cleaning. For integrating-type devices, a thin plastic film is placed over the cups in which the device is held. Thoron decays before it can diffuse through the film but Rn does not.

Detailed methodology on the determination of Radon-222 and radon isotopes in air is given in the Radioanalytical Methods Manual of the Canadian National Uranium Tailings Program (31).

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